Final Report

Title: Exploration of New Principles in Spintronics Based on Spin Hall Insulators

AFOSR/AOARD Reference Number: AOARD 08-4099

AFOSR/AOARD Program Manager: **Gregg H. Jessen**, Ph.D.

Period of Performance: 12 June 2008- 11 June 2010

Submission Date: 11 June 2010

PI: **Yoichi Ando**, Professor, Institute of Scientific and Industrial Research (ISIR), Osaka University

8-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan
Tel: +81-6-6879-8440 / Fax: +81-6-6879-8444

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
13 JUL 2010	**Final**	**12-06-2008 to 11-06-2010**

4. TITLE AND SUBTITLE
Exploration of New Principles in Spintronics Based on Spin Hall Insulators

5a. CONTRACT NUMBER
FA23860814099

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
Yoichi Ando

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Osaka University,8-1 Mohogaoka, Ibaraki,Osaka, Japan,JP,567-0047

8. PERFORMING ORGANIZATION REPORT NUMBER
N/A

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Asian Office of Aerospace Research & Development, (AOARD), Unit 45002, APO, AP, 96338-5002

10. SPONSOR/MONITOR'S ACRONYM(S)
AOARD

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AOARD-084099

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT
This project aimed to explore new avenues of the spintronics to utilize the intrinsic and dissipationless spin current that is expected to flow in spin Hall insulators. During the grant period, the primary objective was to elucidate the basic physics of candidate insulators. In addition, we investigated the nature of quantum spin Hall insulator, which is a quantum-mechanically new sate of matter where an insulating bulk supports an intrinsically metallic, spin-polarized surface state. Our long-term objective is to develop new principles for spintronics devices with minimal energy dissipation, based on the fundamental understanding of those novel materials.

15. SUBJECT TERMS
Physics, Spintronics, Magnetic materials, Quantum Spin Hall insulator

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			
unclassified	**unclassified**	**unclassified**	**Same as Report (SAR)**	**10**	

Objectives:

This project aimed to explore new avenues of the spintronics to utilize the intrinsic and dissipationless spin current that is expected to flow in *spin Hall insulators*. During the grant period, the primary objective was to elucidate the basic physics of candidate insulators. In addition, we investigated the nature of *quantum spin Hall insulator*, which is a quantum-mechanically new sate of matter where an insulating bulk supports an intrinsically metallic, spin-polarized surface state. Our long-term objective is to develop new principles for spintronics devices with minimal energy dissipation, based on the fundamental understanding of those novel materials.

Status of effort:

I. Quantum spin Hall insulator:

1) We have established a method to grow very high quality single crystals of $Bi_{1-x}Sb_x$, which had been theoretically proposed to be a quantum spin Hall insulator for the composition range of $0.07 < x < 0.22$ where the bulk energy gap opens. Taking advantage of our top-notch crystals, we have succeeded in observing two-dimensional Fermi surfaces through quantum oscillations for the first time, which demonstrates that this material indeed has very peculiar surface states [paper #1].
2) We have elucidated the intrinsic spin current in the surface state of $Bi_{1-x}Sb_x$ by using a spin- and angle-resolved photoemission spectroscopy [paper #2].
3) We have discovered unusual angular-dependent magnetoresistance oscillations in $Bi_{1-x}Sb_x$. This unusual phenomenon is a novel manifestation of the surface state [paper #5].
4) Bi_2Se_3 is a "2nd-generation" quantum spin Hall insulator. We have succeeded in growing low-carrier-density Bi_2Se_3 single crystals in the 10^{17} cm^{-3} range. Using our crystals, we have elucidated the anisotropy of its bulk Fermi surface in the low-carrier-density regime [paper #4]. We have also grown low-carrier-density single crystals of Bi_2Te_3, which is another "2nd generation" quantum spin Hall insulator, in the 10^{17} cm^{-3} range. We are currently developing methods to reduce the carrier density down to the 10^{16} cm^{-3} level.
5) We are making efforts to fabricate devices to directly probe the surface spin currents in $Bi_{1-x}Sb_x$ and Bi_2Se_3 through transport experiments. So far, we have identified Al_2O_3 as the most suitable material for the insulation layer, and have elucidated the optimal deposition condition for ferromagnetic contacts.

II. Intrinsic spin Hall insulator:

1) We have successfully grown high-quality single crystals of PbS, a candidate material for the intrinsic spin Hall insulator, and controlled the polarity and the density of its charge carriers.
2) Using the PbS crystals with the carrier concentration in the 10^{17} cm^{-3} range, we discovered an unusual peak effect in the angular-dependent magnetoresistance. The mechanism of this novel effect is currently unknown, but it is most likely associated with the static skin effect and the spin-orbit coupling [paper #3].

Abstract:

I. Research on Quantum spin Hall insulators:

Quantum spin Hall insulator (QSHI), which is also called *topological insulator*, is a new state of matter where an insulating bulk state supports an intrinsically metallic, spin-filtered surface state that is "topologically protected". Recently, QSHI is attracting a lot of attention not only for its fundamental novelty, but also for its potential impact on future spintronics as well as quantum computations. So far three materials, $Bi_{1-x}Sb_x$, Bi_2Se_3, and Bi_2Te_3, have been confirmed to be QSHIs by angle-resolved photoemission spectroscopy (ARPES). During the course of this project, we have grown very high quality single crystals of $Bi_{1-x}Sb_x$ and made the first observation of quantum oscillations originating from the surface state in question, confirming its macroscopic robustness which is a prerequisite for applications. Since it is a common sense in surface physics that conducting surface states are very fragile against disturbances such as adsorption, the fact that a native surface state of a bulk material can be unambiguously seen by transport and magnetization in ambient atmosphere is surprising, and this unusual result immediately signifies the unique properties of the surface states of QSHIs. We have also determined for the first time the complete spin-polarized surface band structure of $Bi_{1-x}Sb_x$, which is a spectroscopic elucidation of the intrinsic spin current in QSHI.

II. Research on Intrinsic spin Hall insulators:

Recently, narrow-gap semiconductors with a strong spin-orbit coupling (SOC) are playing an important role in the condensed matter physics, because topological insulators are found in this family of materials. While the topological insulators are currently under intense investigations, the roles of the strong spin-orbit coupling in the magnetotransport properties are not well understood even in "non-topological" materials. In this context, we have made detailed magnetotransport studies of PbS, which is a non-topological member of the above-mentioned family and is predicted to be an *intrinsic spin Hall insulator* . We have discovered anomalous angular-dependent magnetoresistance which points to the existence of a hitherto-overlooked effect in this family of materials. It is likely to be a manifestation of an intricate interplay between the SOC and the conducting surface layers in the quantum transport regime. This discovery would help establish a general understanding of the magnetotransport in narrow-gap semiconductors with a strong SOC, which is necessary for elucidating the peculiar transport properties of topological insulators.

Personnel Supported:
Alexey A. Taskin, Specially-Appointed Researcher, ISIR, Osaka University (Part-time, Until 31 March 2010)
Kazuma Eto, Graduate Student, ISIR, Osaka University (Part-time)

Publications:

1. A. A. Taskin and Yoichi Ando, "Quantum Oscillations in a Topological Insulator $Bi_{1-x}Sb_x$", Phys. Rev. B **80**, 085303-(1-6) (2009) [Editor's Suggestion].
2. Akinori Nishide, Alexey A. Taskin, Yasuo Takeichi, Taichi Okuda, Akito Kakizaki, Toru Hirahara, Kan Nakatsuji, Fumio Komori, Yoichi Ando, and Iwao Matsuda, "Direct mapping of the spin-filtered surface bands of a three-dimensional quantum spin Hall insulator", Phys. Rev. B (Rapid Communications) **81**, 041309(R)-(1-4) (2010) [Editor's Suggestion].
3. Kazuma Eto, A. A. Taskin, Kouji Segawa, and Yoichi Ando, "Spin-orbit coupling and anomalous angular-dependent magnetoresistance in the quantum transport regime of PbS", Phys. Rev. B (Rapid Communications) **81**, 161202(R)-(1-4) (2010) [Editor's Suggestion].
4. Kazuma Eto, Zhi Ren, A. A. Taskin, Kouji Segawa, and Yoichi Ando, "Angular-dependent oscillations of the magnetoresistance in Bi_2Se_3 due to the three-dimensional bulk Fermi surface", Phys. Rev. B **81**, 195309-(1-5) (2010).
5. A. A. Taskin, Kouji Segawa, Yoichi Ando, "Oscillatory angular dependence of the magnetoresistance in a topological insulator $Bi_{1-x}Sb_x$", submitted to Phys. Rev. Lett. (arXiv:1001.1607).

Interactions:

(a) Participation/presentations at meetings, conferences, seminars, etc.

1. Y. Ando, "Magnetotransport Properties of the Quantum Spin Hall Insulator $Bi_{1-x}Sb_x$", The 5th Symposium on *High Magnetic Field Spin Science in 100 T*, Okayama University, December 12, 2008.
2. K. Eto, T. Kusunose, K. Segawa, and Y. Ando, "Magnetotransport Properties of Carrier-Controlled PbS Single Crystals", The 12th Sanken International Symposium, Osaka University, January 22, 2009.
3. K. Eto, T. Kusunose, K. Segawa, and Y. Ando, "Magnetotransport Properties of Carrier-Controlled PbS", The 64th Annual Meeting of the Physical Society of Japan, Rikkyo University, March 27, 2009.
4. A. Nishide, Y. Takeichi, T. Okuda, A. Kakizaki, K. Nakatsuji, F. Komori, A. Taskin, Y. Ando, and I. Matsuda, "Quantum spin Hall phase in $Bi_{1-x}Sb_x$ studied by high-resolution spin- and angle-resolved photoemission spectroscopy", The 64th Annual Meeting of the Physical Society of Japan, Rikkyo University, March 27, 2009.
5. Y. Ando, "Quantum Oscillations from the Spin-Polarized Surface State of a Topological Insulator", FY2009 Startup Meeting of *High Magnetic Field Spin Science in 100 T*, Institute of Solid State Physics, University of Tokyo, May 22, 2009.

6. Y. Ando, “Transport and Magnetic Studies of the Topological Insulator Bi-Sb”, International Workshop on Novel Topological States in Condensed Matter Physics, University of Hong Kong, June 23, 2009. [**Invited**]

7. Daisuke Hama, Satoshi Sasaki, Alexey Taskin, Nam-Goo Cha, Teruo Kanki, Hidekazu Tanaka and Yoichi Ando, “An Attempt for Observing the Spin-Polarized Charge Current Flowing on the Surface of $Bi_{1-x}Sb_x$ Single Crystal”, Fall Meeting of the Physical Society of Japan, Kumamoto University, September 25, 2009.

8. A. Nishide, Y. Takeichi, A. A. Taskin, Ke He, T. Okuda, K. Nakatsuji, F. Komori, H. Miyazaki, T. Ito, S. Kimura, Y. Ando, A. Kakizaki, and I. Matsuda, “Study of the Sb-doping dependence of the electronic state in the topological insulator $Bi_{1-x}Sb_x$ alloy”, Fall Meeting of the Physical Society of Japan, Kumamoto University, September 27, 2009.

9. Kazuma Eto, Kouji Segawa, Alexey Taskin, Satoshi Sasaki, and Yoichi Ando, “Peculiar quantum oscillations in a candidate spin-Hall-insulator material PbS”, Fall Meeting of the Physical Society of Japan, Kumamoto University, September 27, 2009.

10. Y. Ando, “Anomalous Properties of a Topological Insulator $Bi_{1-x}Sb_x$”, ISSP Workshop on Physical Properties of Dirac electron systems - Recent Research of Graphene and Related Materials, University of Tokyo, October 22, 2009. [**Invited**]

11. Y. Ando, “Anomalous Properties of a Topological Insulator $Bi_{1-x}Sb_x$”, Solid State Engineering Laboratory Seminar, Dept. of Applied Physics, Nagoya University, November 12, 2009. [**Invited**]

12. Y. Ando, “Quantum Oscillations in a Topological Insulator $Bi_{1-x}Sb_x$”, RIKEN Workshop on *Emergent Phenomena of Correlated Materials*, RIKEN, Saitama, December 3, 2009.

13. Kazuma Eto, A. A. Taskin, Kouji Segawa, and Yoichi Ando, “Anomalous magnetic-field-angle dependence of the magnetoresistance in PbS in the quantum transport regime”, RIKEN Workshop on *Emergent Phenomena of Correlated Materials*, RIKEN, Saitama, December 3, 2009.

14. Z. Ren and Yoichi Ando, “Study of the novel superconductivity in Cu-intercalated Bi_2Se_3”, RIKEN Workshop on *Emergent Phenomena of Correlated Materials*, RIKEN, Saitama, December 3, 2009.

15. A. A. Taskin and Yoichi Ando, “Oscillatory Angular Dependence of Magnetoresistance in a Topological Insulator $Bi_{1-x}Sb_x$”, RIKEN Workshop on *Emergent Phenomena of Correlated Materials*, RIKEN, Saitama, December 3, 2009.

16. Y. Ando, “Unusual Transport and Magnetic Properties of a Topological Insulator Bi-Sb”, The 6th International Symposium on *High Magnetic Field Spin Science in 100 T*, Tohoku University, Sendai, December 7, 2009.

17. Y. Ando, “Anomalous Properties of a Topological Insulator Bi-Sb”, Research Meeting of the KAKENHI Priority Area *Physics of New Quantum Phases in Ultraclean Materials*, Wakayama Prefecture Education Center, December 9, 2009. [**Invited**]

18. A. A. Taskin, Kouji Segawa, and Yoichi Ando, “Oscillatory angular dependence of magnetoresistance in a topological insulator $Bi_{1-x}Sb_x$”, ICAM Conference on Exotic Insulating States of Matter, Johns Hopkins University, Baltimore, January 14, 2010.

19. Y. Ando, A. A. Taskin, and Kouji Segawa, “Anomalous Magnetotransport in a Topological Insulator $Bi_{1-x}Sb_x$”, ICAM Conference on Exotic Insulating States of Matter, Johns Hopkins University, Baltimore, January 16, 2010.

20. Y. Ando, “Quantum Oscillations in a Topological Insulator Bi-Sb”, American Physical Society March Meeting, Portland, Oregon, March 15, 2010. [**Invited**]

21. Y. Ando, “Developments of the Study of Topological Insulators: Experiment”, Symposium on Topological Insulators, The 65th Annual Meeting of the Japanese Physical Society, Okayama University, March 22, 2010. [**Invited**]

22. Daisuke Hama, Alexey Taskin, Zhi Ren, Satoshi Sasaki, Kouji Segawa, Nam-Goo Cha, Teruo Kanki, Hidekazu Tanaka, and Yoichi Ando, “An Attempt for Observing the Spin-Polarized Charge Current Flowing on the Surface of a Topological Insulator with Ferromagnetic Contacts”, The 65th Annual Meeting of the Physical Society of Japan, Okayama University, March 23, 2010.
23. Kazuma Eto, Kouji Segawa, Alexey Taskin, and Yoichi Ando, “Possibility of the existence of surface current in the PbS which is a candidate material of spin Hall insulators”, The 65th Annual Meeting of the Physical Society of Japan, Okayama University, March 23, 2010.
24. Y. Ando, “Anomalous Magnetotransport in a Topological Insulator $Bi_{1-x}Sb_x$”, DCMP Seminar, University of Geneva, March 26, 2010. [**Invited**]
25. Y. Ando, “Developments and Prospects of the Studies of Topological Insulators”, Seminar, Yukawa Institute for Theoretical Physics, Kyoto University, May 19, 2010. [**Invited**]

(b) Technology Application: None.

Inventions:

(a) Discoveries, inventions, or patent disclosures: None.

(b) DD Form 882, “Report of Inventions and Subcontractors”: Submitted separately.

Honors/Awards: None.

Archival Documentation: “Spin-orbit coupling and anomalous angular-dependent magnetoresistance in the quantum transport regime of PbS” (attached).

Software and/or Hardware (if they are specified in the contract as part of final deliverables): None.

Spin-Orbit Coupling and Anomalous Angular-Dependent Magnetoresistance in the Quantum Transport Regime of PbS

Kazuma Eto, A. A. Taskin, Kouji Segawa, and Yoichi Ando
Institute of Scientific and Industrial Research, Osaka University, Ibaraki, Osaka 567-0047, Japan

We measured magnetotransport properties of PbS single crystals which exhibit the quantum linear magnetoresistance (MR) as well as the static skin effect that creates a surface layer of additional conductivity. The Shubnikov-de Haas oscillations in the longitudinal MR signify the peculiar role of spin-orbit coupling. In the angular-dependent MR, sharp peaks are observed when the magnetic field is slightly inclined from the longitudinal configuration, which is totally unexpected for a system with nearly spherical Fermi surface and points to an intricate interplay between the spin-orbit coupling and the conducting surface layer in the quantum transport regime.

PACS numbers: 72.20.My, 71.18.+y, 73.25.+i, 75.47.-m

I. INTRODUCTION

Recently, non-trivial consequences of the spin-orbit coupling (SOC) in crystalline solids are a major theme in condensed matter physics[1]. For example, the spin Hall effect is a striking manifestation of the SOC in non-magnetic materials[2], and the SOC in non-centrosymmetric superconductors gives rise to an unconventional order-parameter symmetry[3]. Even more strikingly, it was recognized that a certain class of narrow-gap semiconductors where the energy gap is a product of the SOC are topological insulators, whose valence band structures is characterized by a non-trivial Z_2 topological invariant[4–9]. The three-dimensional topological insulators host helically spin-polarized surface states and are predicted to exhibit various novel phenomena[7–11]. After the discovery of the topological-insulator nature in $Bi_{1-x}Sb_x$, Bi_2Se_3, and Bi_2Te_3 (Refs. 12–15), those three materials are under intense investigations.

In this context, the narrow-gap semiconductor PbS would make a useful comparison, because its energy gap is due to a strong SOC but its valence band structure lends itself to the trivial Z_2 topological class[8]; namely, PbS is a non-topological insulator. Nevertheless, this material may be called an "incipient" spin Hall insulator, since the energy gap of the SOC origin in PbS causes a large Berry phase in the Bloch states and leads to a finite intrinsic spin Hall conductivity $\sigma_{\rm H}^{\rm s}$ even in the insulating state[16]. Therefore, the role of the SOC in the transport properties of this material is worth investigating with the modern understanding.

PbS has a rock salt crystal structure and has a direct energy gap of about 0.3 eV located at the four equivalent L points of the Brillouin zone[17]. Depending on whether S is excessive or deficient, both p- and n-type PbS can be prepared, and in both cases the Fermi surface (FS) is very nearly spherical[17,18]. This material was well studied in the past for its potential in the infrared applications[17]. More recently, PbS is attracting attentions in the photovoltaic community because of the multi-exiton generation[19]. Here, we report our detailed study of the magnetoresistance (MR) in low-carrier-density PbS, focusing on its angular dependence. To our surprise, we observed sharp peaks in the angular-dependent MR in high magnetic fields, which is totally unexpected for a three-dimensional (3D) material with a small spherical FS. Although the exact mechanism of this anomalous behavior is not clear at the moment, our data points to an important role of the SOC in the quantum transport regime. In addition, the formation of a surface layer with additional conductivity due to skipping orbits (called "static skin effect"[20]) appears to be also playing a role in the observed angular dependence. The unexpected angular-dependent MR points to a hitherto-overlooked effect that could become important in the magnetotransport properties of narrow-gap semiconductors with a strong SOC.

II. EXPERIMENTAL DETAILS

High-quality single crystals of PbS were grown by a vapor transport method from a stoichiometric mixture of 99.998% purity Pb and 99.99% purity S. The mixture was sealed in an evacuated quartz tube and was reacted for 5 – 10 h at 980°C. After the reaction, the resulting material was vaporized and transported to the other end of the sealed tube by making a large temperature difference, which worked as a purification stage. The obtained polycrystals were taken out and again sealed in a new evacuated quartz tube for the crystal growth stage: The polycrystal-containing end of the tube was kept at 850°C, and sublimed PbS was transported to the other end kept at 840°C for one week. The obtained single crystals were annealed in sulfur vapor to tune the type and the density of carriers, during which the crystal temperature and the sulfur vapor pressure were controlled independently. We have prepared a series of samples with various carrier density as shown in Fig. 1. In the following we focus on a p-type sample with the carrier density n_h of 4.8×10^{17} cm^{-3}, which was obtained by annealing the crystal at 533°C with the sulfur vapor source kept at 90°C.

The resistivity ρ_{xx} and the Hall resistivity ρ_{yx} were measured simultaneously by using a standard six-probe

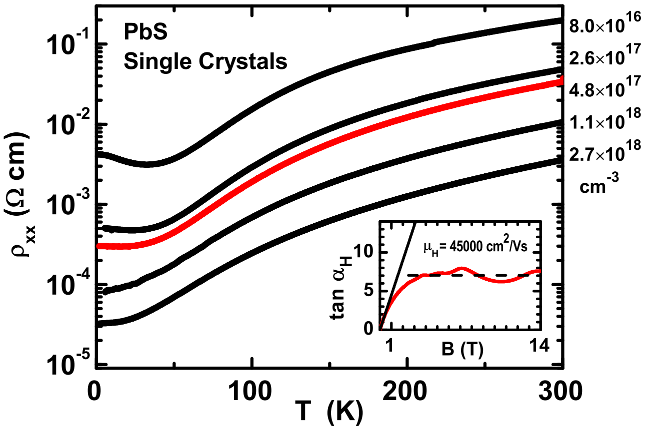

FIG. 1: (Color online) Temperature dependence of ρ_{xx} for a series of p-type PbS single crystals, whose n_h is indicated on the right. Inset shows $\tan\alpha_{\rm H}$ vs B for the 4.8×10^{17} cm^{-3} sample, which exhibits a deviation from the classical linear behavior (shown by the solid straight line) and a saturation above 4 T in the quantum transport regime; the low-field slope is equal to $\mu_{\rm H}$ and gives 4.5×10^4 cm^2/Vs.

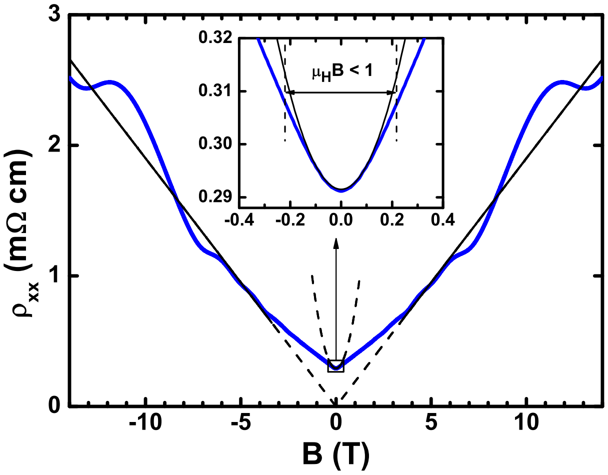

FIG. 2: (Color online) Transverse MR at 1.5 K with B along [001]. The straight lines are the background linear MR, on top of which pronounced SdH oscillations are superimposed. Inset shows the B^2 dependence observed in the low-field classical regime ($\mu_{\rm H}B < 1$) and its fitting (thin solid line).

method on a thin rectangular sample whose top and bottom surfaces were cleaved (001) plane. The current I was always along the [100] direction. The Shubnikov-de Haas (SdH) oscillations were measured by sweeping the magnetic field B between +14 and -14 T for a series of field directions. Continuous rotations of the sample in constant magnetic fields were used for measuring the angular dependence of the MR, in which the direction of the magnetic field B was either [001] → [010] (transverse geometry) or [001] → [100] (transverse to longitudinal geometry).

III. RESULTS AND DISCUSSIONS

A. Magnetoresistance and SdH Oscillations

Figure 2 shows the transverse MR measured at 1.5 K with B along [001]. Pronounced SdH oscillations are clearly seen, and one may notice that the background MR does not show the ordinary B^2 dependence. This is because the range of the weak-field regime ($\mu_{\rm H}B < 1$), where the B^2 dependence is observed, is extremely narrow in our sample, as shown in the inset of Fig. 2 ($\mu_{\rm H}$ is the Hall mobility). It is worth noting that our sample shows a nearly-B-linear background MR in the strong-field regime ($\mu_{\rm H}B > 1$), rather than a tendency to saturation which is usually observed in metals. This high-field behavior is the so-called "quantum linear MR" proposed by Abrikosov[21]. Such a behavior is expected in the quantum transport regime where the condition $n_h < (eB/\hbar c)^{2/3}$ is satisfied[21], and in our sample this regime is realized for $B > 4$ T. In this sense, our PbS presents a straightforward realization of the quantum linear MR and is different from $Ag_{2+\delta}Se$ or $Ag_{2+\delta}Te$ where the linear MR is observed down to very low field[21].

The SdH oscillations measured in the transverse ($B \perp I$) and longitudinal ($B \parallel I$) MR are presented in Fig. 3 by plotting $d\rho_{xx}/dB$ vs $1/B$. One can see that we are resolving the spin-splitting of the Landau levels in high magnetic fields and that the crossing of the 0^- state by the Fermi level is observed (see the lower left inset of Fig. 3 for the Landau level diagram); this means that all the electrons are in the 0^+ state in the highest field (14 T) and hence the system is in the quantum limit. Also, one may notice that the amplitude of some particular peaks, 0^-, 1^+, and 2^+, are significantly diminished in the longitudinal configuration ($B \parallel I$) compared to the transverse one where all the peaks are well developed. Such a behavior was previously observed in $Hg_{1-x}Cd_xTe$ (Ref. 22) and in $Pb_{1-x}Sn_xTe$ (Ref. 23), and was elucidated to be due to some selection rules[22] imposed by the SOC which prohibits scattering between certain Landau sublevels in the longitudinal configuration. Besides this peculiar difference, the peak positions are almost the same for the two field directions, which is because the FS in PbS is nearly spherical[17,18]. Also, the cyclotron mass m_c extracted from the temperature dependence of the SdH amplitude (lower right inset) using the Lifshitz-Kosevich formula[24] is identical for the two directions.

The carrier density n_h calculated from the FS volume seen by the SdH oscillations is 4.8×10^{17} cm^{-3}, which agrees well with the value of $n_h = 4.6\times10^{17}$ cm^{-3} obtained from the high-field Hall coefficient $R_{H\infty}$. It is worth noting that the Hall mobility $\mu_{\rm H}$ calculated from $R_{H\infty}$ and ρ_{xx} at 0 T is 4.5×10^4 cm^2/Vs, while the mobility $\mu_{\rm SdH}$ obtained from the SdH oscillations is only 3.8×10^3 cm^2/Vs (Ref. 25). This discrepancy is essentially due to the fact that ρ_{xx} and the Dingle temperature T_D (smearing factor in the SdH effect[24]) are determined

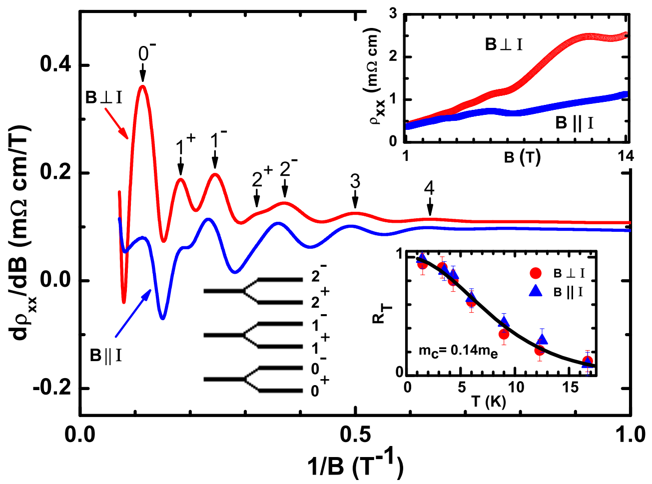

FIG. 3: (Color online) SdH oscillations in the transverse ($B \perp I$) and longitudinal ($B \parallel I$) MR at 1.5 K. Upper inset shows the raw data, and the main panel presents $d\rho_{xx}/dB$ vs $1/B$. In the "$B \parallel I$" measurement, the magnetic field was 8° off from the exactly parallel direction. Lower left inset shows the schematic Landau level diagram. Lower right inset shows the temperature dependence of the normalized SdH amplitude, which yields $m_c = 0.14 m_e$ (m_e is the free electron mass).

by different scattering processes; namely, ρ_{xx} is primarily determined by the backward scattering, while T_D is sensitive to both forward and backward scattering[24,26]. Apparently, only small-angle scatterings are relevant in high-mobility PbS, which leads to the 12 times difference between $\mu_{\rm H}$ and $\mu_{\rm SdH}$.

B. Angular Dependence of Magnetoresistance

Now let us present the most surprising result. The angular-dependent MR for the transverse-to-longitudinal rotation (I was along [100] and B was rotated from [001] toward [100]) is shown in Fig. 4(a), and the corresponding angular dependence of ρ_{yx} is shown in Fig. 4(b) (the magnetic-field angle θ is measured from [001]). In Fig. 4(a), pronounced peaks are observed when θ is near 90°, that is, when the magnetic field is slightly inclined from the longitudinal configuration. The θ dependence of ρ_{yx} also shows a feature near 90°, which can be more easily seen in the upper inset of Fig. 4(b) where the deviation of the measured $\rho_{yx}(\theta)$ from a smooth $\cos\theta$ dependence is plotted together with the $\rho_{xx}(\theta)$ data (which is multiplied by 0.3). One can easily see in this inset that the sharp peak occurs in both ρ_{xx} and ρ_{yx} at the same θ; in addition, $\rho_{yx}(\theta)$ apparently shows periodic oscillations in a wide range of θ, whose relation to the sharp peak is not obvious. In any case, given that the FS in PbS is nearly spherical and that they cannot give rise to any open orbit, such a sharp peak in the angular-dependent MR is totally unexpected.

To gain insight into the origin of the unexpected peak

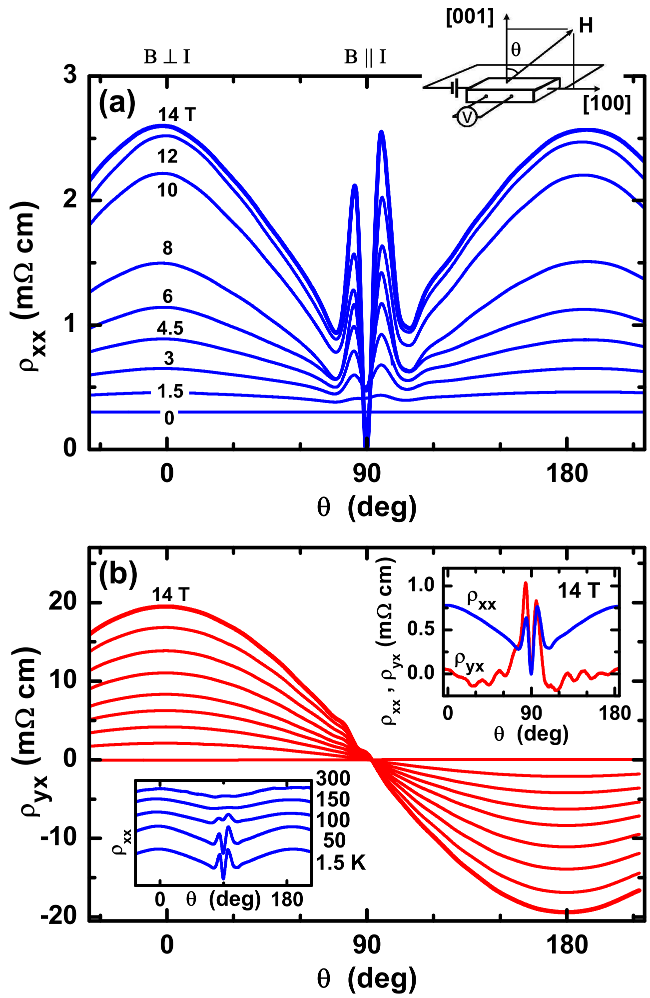

FIG. 4: (Color online) Angular-dependence of (a) ρ_{xx} and (b) ρ_{yx} for the transverse-to-longitudinal rotation (top inset shows the geometry). In panel (b), the upper inset compares the deviation of $\rho_{yx}(\theta)$ from the $\cos\theta$ dependence to $\rho_{xx}(\theta)$ multiplied by 0.3, while the lower inset shows how the anomalous peaks weaken with temperature (data are vertically shifted for clarity).

in $\rho_{xx}(\theta)$, the angular-dependent MR data in a different rotation plane is useful. Figure 5 shows such data for the transverse rotation (I was along [100] and B was rotated from [001] toward [010]). As one can see in Fig. 5, there is no sharp peak in this configuration, which immediately indicates that the unexpected peaks are peculiar to the near-longitudinal configuration. Besides the absence of the sharp peaks, there is a notable feature in Fig. 5: Since PbS has a cubic symmetry, the MR for $\theta = 0°$ (B along [001]) and 90° (B along [010]) should be the same, since [001] and [010] are crystallographically identical and the measurement configuration is both transverse. However, the actual data in Fig. 5 indicates that they are different, which suggests that there must be some additional factor which affects the resistivity in magnetic field. In the past, similar anisotropy was ob-

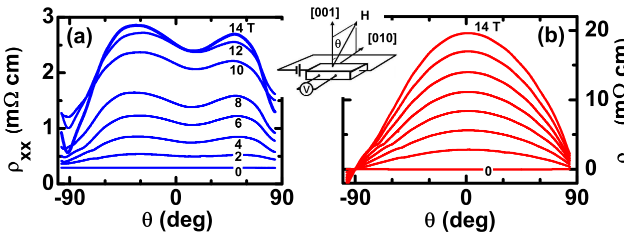

FIG. 5: (Color online) Angular-dependence of (a) ρ_{xx} and (b) ρ_{yx} for the transverse rotation (inset shows the geometry). The ρ_{xx} data shown here are after removing the admixture of the ρ_{yx} component in the ρ_{xx} measurement.

served in clean specimens of low-carrier-density materials such as Bi (Ref. 27) and Sb (Ref. 28), and was explained in terms of the static skin effect[20], that is, the formation of a surface layer of additional conductivity due to skipping orbits when the magnetic field is nearly parallel to a specular surface. In this regard, since the top and bottom surfaces of our sample were cleaved (001) plane, it is understandable that the static skin effect creates a surface conduction layer near the specular (001) surface for the magnetic field along [010], leading to a reduced resistivity.

An additional factor to consider regarding the MR anisotropy in the present case is the SOC which diminishes some of the peaks in the SdH oscillations for $B \parallel I$. In fact, as one can infer in the upper inset of Fig. 3, the change in the SdH oscillations due to the SOC is partly responsible for the difference in MR between $B \perp I$ and $B \parallel I$. Another factor to consider is the crossover between classical and quantum transport regimes: As we already discussed, the quantum regime is arrived above 4 T in the transverse configuration. (This crossover can also be seen in the B dependence of $\tan \alpha_{\rm H}$, which is linear in B in the classical regime but saturates in the quantum regime[29], see Fig. 1 inset.) On the other hand, in the longitudinal configuration, the electron motion along the current direction is *not* quantized, and therefore ρ_{xx} for $B \parallel I$ is always "classical". This means that in our measurement in high magnetic fields, there is a crossover from the classical to the quantum regime when the configuration changes from longitudinal to transverse. Since the MR behavior is different in the two regimes, this crossover must be partly responsible for the observed MR anisotropy.

Although we have not been able to elucidate the mechanism for the sharp peaks in the angular-dependent MR shown in Fig. 4(a), we can see that there are three factors that are likely to participate in this phenomenon: (i) the SOC which diminishes some of the peaks in the SdH oscillations for $B \parallel I$, (ii) the static skin effect which creates a conducting surface layer, and (iii) the crossover between classical and quantum transport regimes. It is useful to note that the sharp peaks weaken only gradually with increasing temperature and are still observable at 100 K [lower inset of Fig. 4(b)], while the SdH oscillations disappear above ~20 K (lower right inset of Fig. 3); this suggests that the sharp peak is not directly related to quantum oscillations. We also note that the SOC is expected to affect not only the SdH oscillations but also the static skin effect when the magnetic field is inclined from the surface, because in such a configuration the surface reflection of an electrons necessarily involves a transition to a different Landau level[20], and the same selection rules imposed by the SOC as those in the SdH case[22] would apply. We expect that the anomalous angular-dependent MR is a result of an intricate interplay between the above three factors.

IV. CONCLUSIONS

In conclusion, we have observed sharp peaks in the angular-dependent MR in PbS when the magnetic field is slightly inclined from the longitudinal ($B \parallel I$) configuration, which is totally unexpected for a low-carrier-density system with nearly spherical Fermi surface. While the mechanism of this peak is to be elucidated in future, we show that the spin-orbit coupling, the static skin effect, and the crossover between classical and quantum transport regimes, are all important in the magnetotransport properties of PbS. This unusual phenomenon would help establish a general understanding of the magnetotransport in narrow-gap semiconductors with a strong SOC, which is important in elucidating the transport properties of topological insulators.

Acknowledgments

This work was supported by JSPS (KAKENHI 19340078 and 2003004) and AFOSR (AOARD-08-4099). We thank H. D. Drew and V. Yakovenko for discussions.

[1] N. Nagaosa, J. Phys. Soc. Jpn. **77**, 031010 (2008).
[2] S. Murakami, Adv. in Solid State Phys. **45**, 197 (2005).
[3] S. Fujimoto, J. Phys. Soc. Jpn. **76**, 051008 (2007).
[4] C. L. Kane and E. J. Mele, Phys. Rev. Lett. **95**, 146802 (2005).
[5] B. A. Bernevig, T. L. Hughes, and S.-C. Zhang, Science **314**, 1757 (2006).
[6] J. E. Moore and L. Balents, Phys. Rev. B **75**, 121306(R) (2007).
[7] L. Fu, C. L. Kane, and E. J. Mele, Phys. Rev. Lett. **98**, 106803 (2007).
[8] L. Fu and C. L. Kane, Phys. Rev. B **76**, 045302 (2007).
[9] X.-L. Qi, T. L. Hughes, and S.-C. Zhang, Phys. Rev. B **78**, 195424 (2008).

[10] L. Fu and C. L. Kane, Phys. Rev. Lett. **100**, 096407 (2008).
[11] X.-L. Qi, R. Li, J. Zang, and S.-C. Zhang, Science **323**, 1184 (2009).
[12] D. Hsieh, D. Qian, L. Wray, Y. Xia, Y. S. Hor, R. J. Cava, and M. Z. Hasan, Nature **452**, 970 (2008).
[13] H.-J. Zhang, C.-X. Liu, X.-L. Qi, X. Dai, Z. Fang, and S.-C. Zhang, Nat. Phys. **5**, 438 (2009).
[14] Y. Xia, D. Qian, D. Hsieh, L. Wray, A. Pal, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, and M. Z. Hasan, Nat. Phys. **5**, 398 (2009).
[15] Y. L. Chen, J. G. Analytis, J.-H. Chu, Z. K. Liu, S.-K. Mo, X. L. Qi, H. J. Zhang, D. H. Lu, X. Dai, Z. Fang, S. C. Zhang, I. R. Fisher, Z. Hussain, and Z.-X. Shen, Science **325**, 178 (2009).
[16] S. Murakami, N. Nagaosa, and S.-C. Zhang, Phys. Rev. Lett. **93**, 156804 (2004).
[17] R. Dalven, Infrared Phys. **9**, 141 (1969).
[18] P. J. Stiles, E. Burstein, and D. N. Langenberg, J. Appl. Phys. **32**, 2174 (1961).
[19] J. J. H. Pijpers, R. Ulbricht, K. J. Tielrooij, A. Osherov, Y. Golan, C. Delerue, G. Allan, and M. Bonn, Nat. Phys. **5**, 811 (2009).
[20] V. G. Peschanskii and M. Y. Azbel', Soviet Phys. JETP **28**, 1045 (1969).
[21] A. A. Abrikosov, J. Phys. A: Math. Gen. **36**, 9119 (2003).
[22] K. Suizu and S. Narita, Phys. Lett. A **43**, 353 (1973).
[23] S. Narita and Y. Takafuji, Solid State Commun. **20**, 357 (1976).
[24] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, 1984).
[25] Analysis of the SdH data yields $T_D = 4$ K, which gives the scattering time 3×10^{-13} s. Using $m_c = 0.14m_e$, we obtain $\mu_{\rm SdH} = 3.8\times10^3$ cm^2/Vs. The observed SdH frequency of 7.7 T corresponds to $k_F = 1.53\times10^6$ cm^{-1}, which gives E_F = 6.4 meV, $v_F = 1.26\times10^7$ cm/s, and $\ell_{\rm SdH} = 3.8\times10^{-6}$ cm. This is contrasted to the transport mean free path $\ell_{\rm tr}$ = 4.5×10^{-5} cm.
[26] S. Das Sarma and F. Stern, Phys. Rev. B **32**, 8442 (1985).
[27] Y. A. Bogod, V. V. Eremenko, and L. K. Chubova, Soviet Phys. JETP **29**, 17 (1969).
[28] Y. A. Bogod and V. B. Krasovitskii, Soviet Phys. JETP **36**, 544 (1973).
[29] In the quantum regime, both ρ_{xx} and ρ_{yx} becomes linear in B, leading to a constant $\tan\alpha_{\rm H}$.